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**This paper was prepared for submittal to the
28th Annual Symposium on Optical Materials
for High Power Lasers '96
Boulder, Colorado
October 7-9, 1996**

February 12, 1997



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Investigation of Damage in KDP Using Light Scattering Techniques

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Abstract

Interest in producing high damage threshold KH_2PO_4 (KDP) and $(\text{D}_x\text{H}_{1-x})_2\text{PO}_4$ (DKDP) (also called KD*P) for frequency conversion and optical switching applications is driven by the requirements of the National Ignition Facility (NIF) at the Lawrence Livermore National Laboratory (LLNL). At present only the best crystals meet the NIF system requirements at the third harmonic (351nm) and only after a laser conditioning process. Neither the mechanism for damage in bulk KDP nor the mechanism for conditioning is understood. As part of a development effort to increase the damage thresholds of KDP and DKDP, we have been developing techniques to pinpoint the locations where damage will initiate in the bulk material. After we find these locations we will use other measurement techniques to determine how these locations differ from the other surrounding material and why they cause damage. This will allow crystal growers to focus their efforts to improve damage thresholds.

Historically, damage thresholds have increased it is believed as a consequence of increased purity of the growth solution and through the use of constant filtration during the growth process.¹ As a result we believe that damage is caused by defects in the crystals and have conducted a series of experiments using light scatter to locate these defects and to determine when and where damage occurs. In this paper we present results which show a low correlation between light scatter from bulk defects in KDP and the initiation sites for damage.

We have also studied the effects of thermal conditioning on light scatter, strain induced birefringence and damage threshold. We have seen evidence that regions of high strain also exhibit lower damage threshold than the surrounding lower strain material. When thermally conditioned, these crystals show a decrease in some of the strong linear scattering features and a decrease in the strain birefringence while the damage threshold in these regions increased to that of the surrounding bulk material.

Keywords: KDP, DKDP, bulk damage, optical scatter, thermal conditioning, strain birefringence

1. Introduction

LLNL has historically used KDP grown by conventional slow growth processes (using low supersaturation) for frequency conversion of the large solid state laser systems. In addition to using KDP for the second harmonic converter and DKDP for the third harmonic converter, KDP will be used as the electro-optic material in a large aperture Pockels cell used in a multipass architecture employed on the NIF. Researchers at LLNL have been developing a rapid growth process (using high supersaturation) for growing KDP that is capable of growth rates 10 times that of conventional techniques.² This technique is capable of producing crystals with excellent optical properties at sizes that are comparable to conventionally grown crystals.

Table 1 shows the NIF damage threshold requirements at the use wavelength for a 3ns pulsewidth for each location where KDP and DKDP will be used. The redline values presented are the most stringent requirements for the crystals based on the most demanding design scenario for the NIF. At present only the best crystals meet the NIF system requirements at the third harmonic (351nm) and only after a laser conditioning process. In this position, DKDP is used to suppress stimulated Raman scatter.³

Component / Material	Wavelength (nm)	NIF Redline Fluence (J/cm ²)
Pockels Cell KDP	1053	11.7
Second Harmonic Converter KDP	1053	18.1
Third Harmonic Converter DKDP	351	14.3

Growth Process	Wavelength (nm)	S:1 Threshold (J/cm ²)	R:1 Threshold (J/cm ²)
Conventional Growth KDP	1064	34	43
Conventional Growth DKDP	355	10	20
Rapid Growth KDP	355	6	10

Table 1

Damage threshold values were measured at LLNL using small spot measurements and have been scaled to 3ns values using ^{0.5}. Our test procedures were done using standard methods with S on 1 results indicating the standard test of 600 shots at the same fluence on each site (unconditioned test) and R on 1 results indicating a ramped fluence from zero to the final test fluence over 300 shots and then remaining at the final test fluence for an additional 300 shots (conditioned test). Damage was detected using the standard method of 100X darkfield microscopy. Thresholds for conventionally grown crystals represent values from witness samples from crystals which have been used on Beamlet and are some of the highest values measured on KDP and DKDP at LLNL.⁴ Rapid growth thresholds are average values for crystals grown recently for which the starting salt was carefully monitored. We have measured R on 1 damage thresholds for fast grown crystals at 355nm and 3ns as high as 13.9J/cm².

2. Scatter

Proceeding with the belief that damage was caused by inclusions, a diagnostic was implemented to study side scattered light from inclusions and strained regions in KDP. This diagnostic was fielded on the ZEUS laser damage facility at LLNL, see Figure 1. ZEUS is a high fluence damage test facility that operates at 10Hz and is capable of delivering fluences of 100J/cm² at 1064nm and 50J/cm² at 355nm in 8ns with a 1mm diameter spot at 1/e². The diagnostic beam which was derived from an intracavity doubled YAG laser was aligned nearly collinear to the high fluence test beam in order to illuminate the same volume as the high fluence pulse. A high dynamic range (14 bit) thermoelectric cooled CCD camera allowed any defect in the bulk material that scattered light in the volume irradiated with the high fluence pulse to be studied immediately before and after the pulse. Since the diagnostic viewed 90° side scattered light, any defects would show up as a bright region in a darkfield background. This apparatus has also been referred to in the literature as an ultramicroscope.⁵ Overlap of the two beams was accomplished using ZEUS to damage a silver mirror that replaced the KDP sample and aligning the probe laser to the damage spot using a standard CCD camera. The sample was mounted on a precision stage that allowed X Y translation so the beam could be moved to any point on the sample. The stage also allowed the sample to be rotated into the beamline for damage testing and viewing with the high resolution camera or rotated under a 100 X darkfield microscope for viewing damage using the standard backlighting techniques.⁶

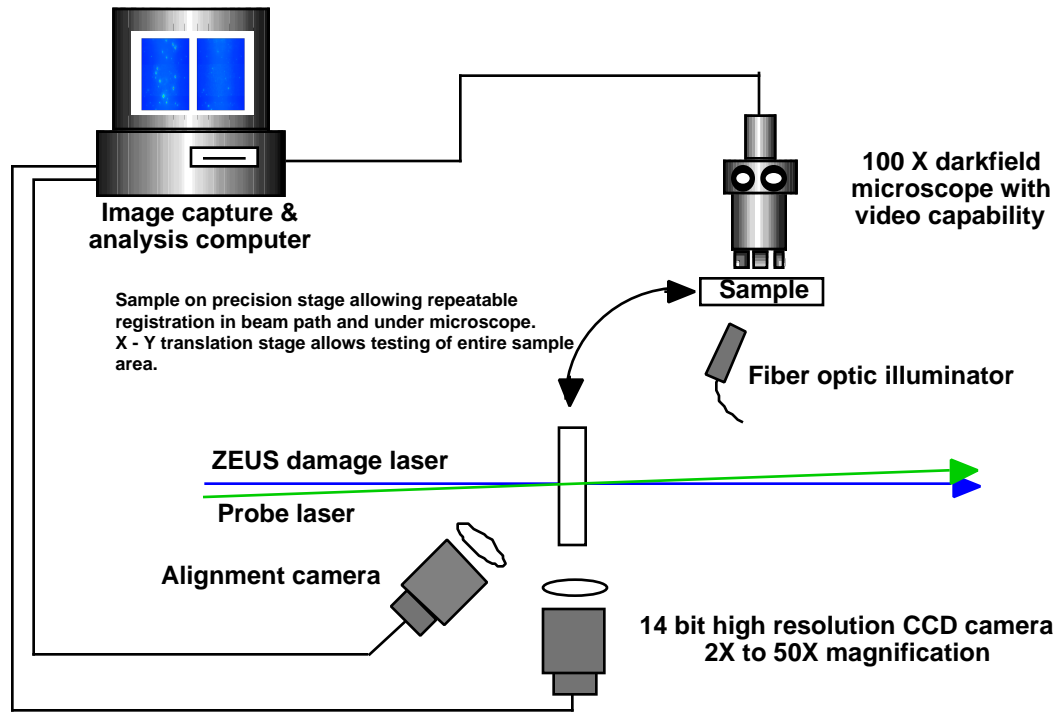


Figure 1. Experimental Layout

Figure 2 shows the three types of scatter signals that were observed with our scatter diagnostic: point, line, and Rayleigh. In each of these images we are looking through a polished edge of the sample with the beam traveling from top to bottom. The field of view has been limited to preclude surface scatter since it is much more intense than bulk scatter and would saturate the camera. The images presented in this paper have been individually scaled to highlight important features.

Figure 2a is a typical example of what we call point scatter. Until recently we were never able to see any topography to these defects even under high magnification. Two boules were grown recently where in the first case the starting salt was dissolved into solution with no filtration of the solution prior to growth and in the second case the solution was initially filtered to $0.02\mu\text{m}$ and then Lucite and Teflon particulates were added intentionally. In each of these boules a very small number of the point scatterers exhibited topography when viewed with a long working distance optical microscope using various lighting techniques. These were the first crystals in which we were able to observe this topography. These recent results indicate that particulates can indeed be incorporated in the crystals during growth.

Damage tests were conducted on various crystals using the scatter diagnostic where images were taken immediately before and after the sample was irradiated with a high fluence pulse to determine if the point scatterers were the initiation sites for damage. Figure 3 shows the three results that we have seen. First, on occasion, a preexisting scatter site would serve as the initiation point for laser damage but these occurrences were rare. Secondly, damage most often did not initiate at a preexisting scatter site. A final observation was that the scatter signal from preexisting scatter sites often decreased dramatically when irradiated with high fluence pulses from ZEUS.

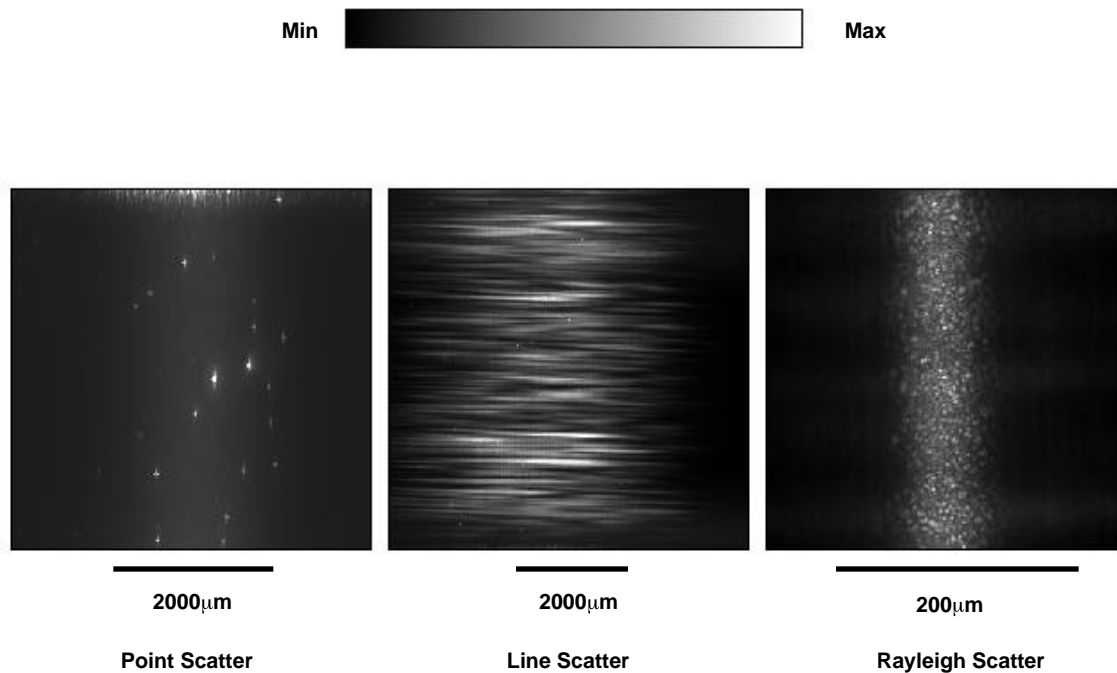


Figure 2. Three types of scatter a) point scatter, b) line scatter, c) Rayleigh scatter.

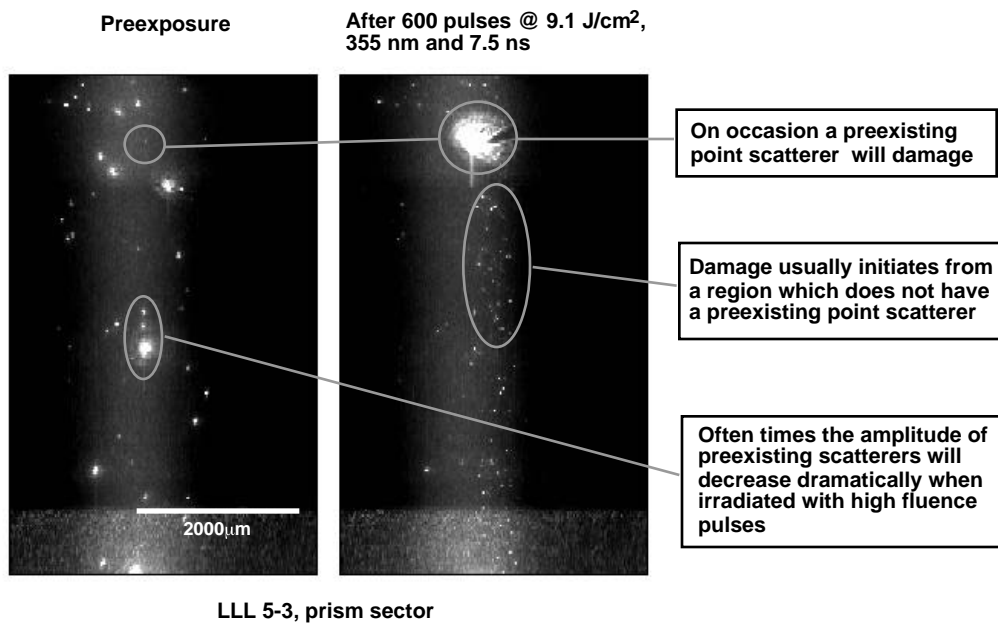


Figure 3. Before and after damage images show that one very dim preexisting site damages, numerous other damage sites initiate in regions that show no preexisting sites and finally, that the scatter signal from some preexisting sites decreases dramatically.

During our study of various rapid growth and slow growth crystals we have seen a dramatic difference in the number of point scatterers and their amplitude between crystals that incorporated constant filtration and those that did not. Recently, constant filtration has been implemented in the rapid growth process at LLNL. Figure 4 shows the dramatic difference between a rapid growth crystal utilizing constant filtration and a rapid growth crystal that didn't. It has been reported that constant filtration of the growth solution during growth has resulted in higher damage thresholds.¹ This result would suggest that defects in the crystals which cause point scattering are generated by contaminants in the growth solution which are filtered out with the constant filtration process. Recent test results on two rapid growth crystals suggest that while constant filtration may impact the damage threshold it is by no means the entire answer. We tested two crystals with very similar salt and growth parameters, the main variable being that one was grown without using the constant filtration process and one with the constant filtration process. The results from the damage threshold measurements were virtually the same. The fact that we do not, in general, see point scatterers initiate damage and that damage tests with rapid grown crystals show no improvement in damage threshold with the use of constant filtration suggests that constant filtration does not have as strong an impact on raising damage thresholds as previously thought. At present we have only a loose historical correlation linking constant filtration to improved damage thresholds. It should be noted that while we typically do not find that point scatterers initiate damage, occasionally some do. This leads us to conclude that, although they are not the primary source of damage, point scatterers are potentially bad and should be eliminated.

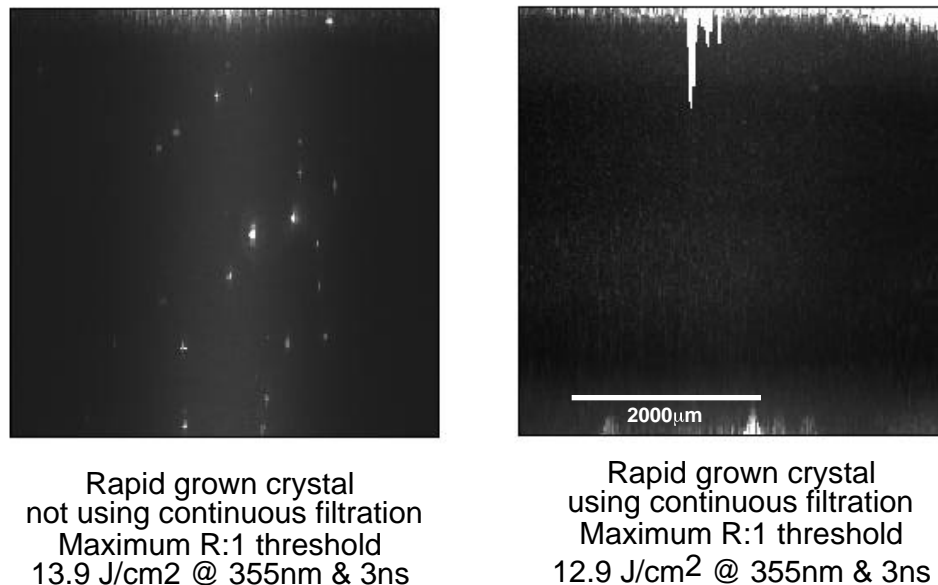


Figure 4. Example of rapid grown crystals grown with and without constant filtration.

The result that the point scatterers were not the initiation sites for damage was quite unexpected and we performed numerous tests to better understand our scatter diagnostic and the nature of point scatterers. To answer the question as to whether we were seeing all scatter sites and whether there was a preferred illumination or viewing direction, a crystal was fabricated with many polished facets allowing us to illuminate and view scatter from various angles. These tests showed that we were indeed seeing all of the point scatterers but that the amplitude depended on the viewing direction. We also found that the intensity of the point scatterers depended on the polarization of the probe laser and the illumination angle. The polarization content of the scatter depended on the individual scattering site. A further complicating factor was that when the scatterers were viewed in a direction with birefringence for the light emanating from the

scatterer, two distinct focal locations were seen which tended to confuse the image. We also looked at forward scattered light since a spherical Mie scatterer should have the largest amplitude of scattered light in the forward direction. We found that the scattered intensity was larger in the forward direction but the images were more confused due to the birefringence of the crystal. The images did not show different or any more scatterers than with 90° scattered light

Our scatter diagnostic using a highly directional-monochromatic illumination source and viewing 90° side scattered light may not display the true shape of a scatter site. Taking the most simple case, we viewed a bubble in a nonbirefringent medium (fused silica) and found that the bubble appeared as if it were two point sources and was not visually recognizable as a bubble. This result is due to the high directionality of the illumination source. Interference effects due to the monochromatic laser source can also confuse the images.

None-the-less the scatter diagnostic has proven to be very sensitive in detecting damage in the bulk material. Damage is manifested as a small dim point source for very slight damage to a large bright region for massive damage. This technique has proven to be much more sensitive in detecting damage or changes in bulk KDP than the standard 100X optical microscope utilizing darkfield techniques that have been used for years in damage tests for bulk material at LLNL. We typically always see damage with the scatter diagnostic before we see it using the optical microscope and when we do begin to see damage using the optical microscope the damage sites are very apparent using the scatter diagnostic. With this and other improved techniques being developed, we are being forced to now think about functional damage thresholds for components to be used for the NIF. As we become able to see smaller and smaller changes to components when irradiated with high fluence pulses, we are able to detect permanent changes to materials and coatings that may have no bearing on the ultimate performance of the NIF.⁷

After finding that we gained little insight into where damage will initiate studying point scatterers we began studying another type of scatter seen in the crystals, linear scatter. Figure 2b is an example of linear scatter features seen in KDP. These features are parallel to the growth front in the crystal and show differences in lattice parameters in the crystal. These differences could be due to impurities or strain which would translate into a local index variation. In order to avoid interference effects from scatter due to growth stria at various layers in the crystal a sheet beam, 2000μm X 150μm, is used to interrogate the sample. The visibility of the line features is sensitive to tip and tilt of the crystal and move as expected as the beam is translated in the crystal. The scatter signal we observe is dependent on the location relative to the direction in which the scatter is viewed. A sample in which the faces are normal to the Z axis and containing the sector boundaries for the pyramidal faces is shown in Figure 5. The resulting scatter signals for three beam locations for an observer peering in from the edge of the crystal are shown. The growth front is oriented such that the observer detects direct scattering from stria in image 5a. Image 5b also shows direct scattering from the pyramidal sector closest to the observer but in addition the sector boundary is clearly visible as a hard edge to the line scattering. In image 5c the line scatter is not visible to the observer because they are not positioned to see direct scatter from growth stria for the pyramidal sector in the lower section of the crystal.

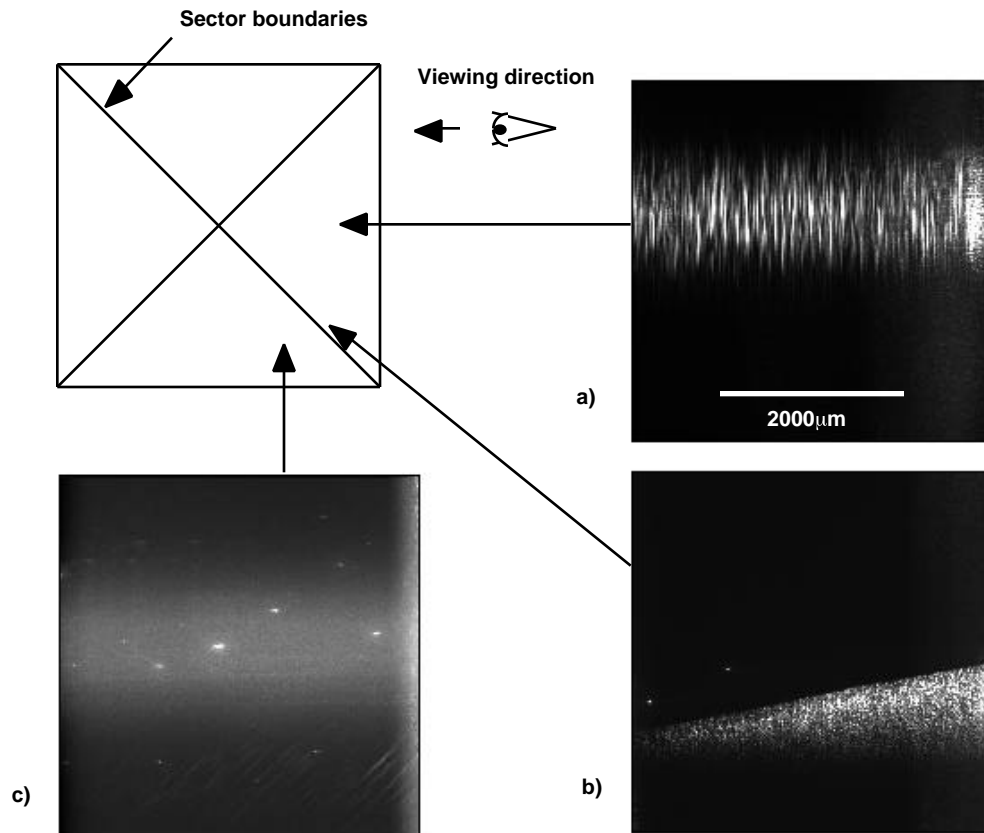


Figure 5. Scatter images for a z cut plate. Beam is traveling into the paper and observer is looking through the edge of the crystal as shown.

A substantial difference was noted in the intensity of the linear features both from crystal to crystal and within the same crystal. On average the rapid grown crystals exhibited linear features with larger intensities than conventionally grown crystals. Crystal 328, a conventionally grown crystal with very high damage threshold displayed linear features whose density and amplitude was very low.

A series of measurements were conducted to determine if there was a correlation between these linear features and damage. The first set of measurements used the scatter diagnostic to look at the linear features and their relation to where damage initiates. The results were mixed. In some cases the damage seemed to occur on or very near a line feature, as shown in Figure 6, and in other cases it did not. Samples with a high density of line features such as in Figure 2b made it very difficult to draw conclusions about whether damage occurred on line features because no matter where the damage occurred one could make the argument either way. Most of our useful tests were performed on samples with a low density of line features.

The second series of tests was conducted on one of the first rapid grown crystals to utilize the constant filtration process. This crystal had regions where the amplitude of the scatter signal from the line features varied by three orders of magnitude. This crystal presented us with the opportunity to study whether the amplitude of the line features had any correlation to the damage threshold. R on 1 and S on 1 tests were performed at numerous test sites in regions covering all amplitudes of line features. We found that there was no correlation between amplitude of scatter signal and damage threshold.

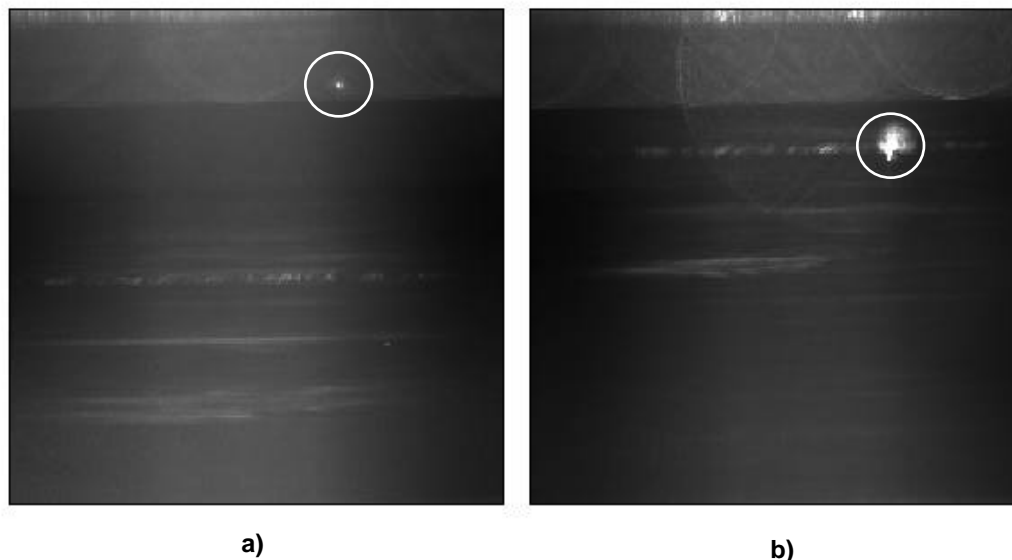


Figure 6. Images showing damage occurring not on a line feature (a), and on a line feature (b).

Figure 3c is an example of Rayleigh scattering seen with our scatter diagnostic. These higher magnification images were acquired in the same way as other scatter images except we employed microscope optics with a long working distance as the imaging optics for the high dynamic range CCD camera. Damage tests have not yet been performed to determine if a link exists between Rayleigh scatter and the initiation sites for damage.

3. Thermal Conditioning

We have begun to look at the effects of thermal conditioning on light scatter, strain, and damage threshold in KDP and DKDP. A rapid grown crystal, LLNL 5, was studied extensively because it had a region that exhibited high strain, high amplitude of light scatter, and low damage threshold. We measured the strain induced birefringence in the crystal using a circular polarimeter developed at LLNL⁸ before and after a thermal conditioning process. Figure 7 shows the strain map of a plate cut from LLNL 5. Plotted is the signal which has been depolarized by the crystal due to strain induced birefringence. The high strain region was found to correspond to the prism-pyramidal sector boundary by comparing the depolarization map to UV absorption data for the crystal. A witness sample was cut from the larger plate and was thermally conditioned for 72 hours at 160°C. Notice that the depolarization decreased from 1% before conditioning to 0.26% after conditioning and that the low loss regions remained relatively unchanged. This is consistent with observations several years ago where we measured the change in strain induced birefringence for several low loss conventionally grown crystals and found the loss to be essentially unchanged after thermal conditioning.

When LLNL 5 was damage tested it was found that the strained region near the sector boundary, which also showed high intensity of light scatter due to the strain, always damaged at fluences well below the threshold of the surrounding material. When the crystal was thermally conditioned the damage threshold in the strained region increased to that of the surrounding material. A positive effect of thermal conditioning on damage thresholds has been reported previously^{9,10} but in these experiments we have linked highly strained regions with low damage thresholds and have seen that thermal conditioning decreases strain and increases

damage threshold. We also saw that the high intensity scatter in the strained region decreased to that of the surrounding material after thermal conditioning.

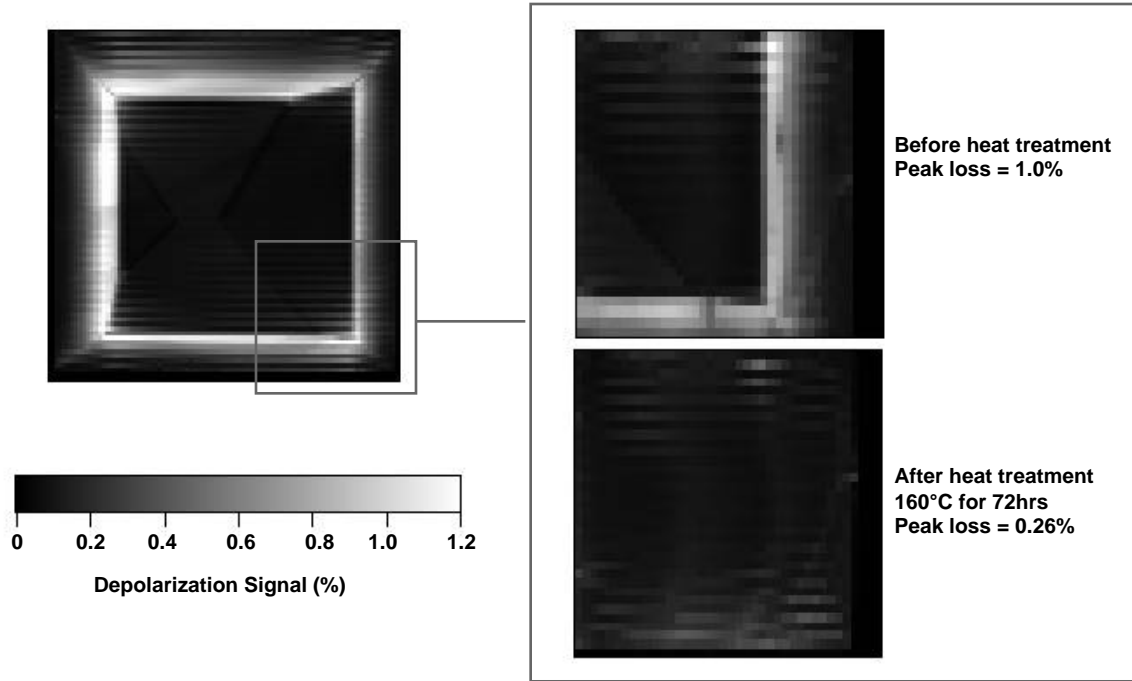


Figure 7. Strain map of LLNL 5 before and after thermal conditioning.

Thermal conditioning decreases the amplitude of linear features with large amplitudes but has little observable effect on line features with low amplitude. We have seen no evidence that thermal conditioning has any effect on point scatterers. Figure 8 shows an example of linear features at the same location in a KDP crystal before and after thermal conditioning (seven days at 160° C). Each image and a vertical lineout immediately to its right are scaled the same to illustrate that the large amplitude features decrease in intensity whereas the much smaller amplitude linear features remain about the same. Figure 9 shows the dependence of the amplitude of the linear features on thermal conditioning. Plotted are peak scatter signals versus annealing time after normalizing for exposure time and laser power for five different locations in the crystal. This result shows that most of the benefit is arrived at in 8 to 10 days.

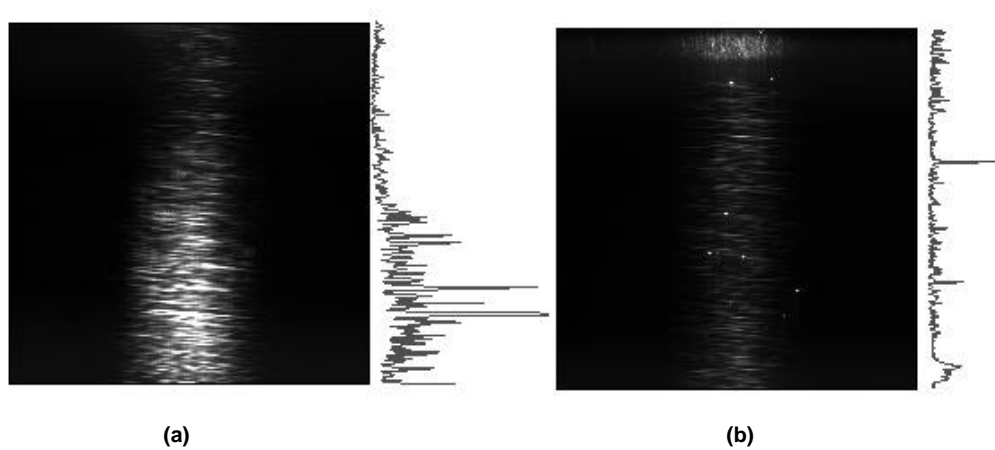


Figure 8. Linear features before and after thermal conditioning. Vertical line outs for each image are shown to the right of each image.

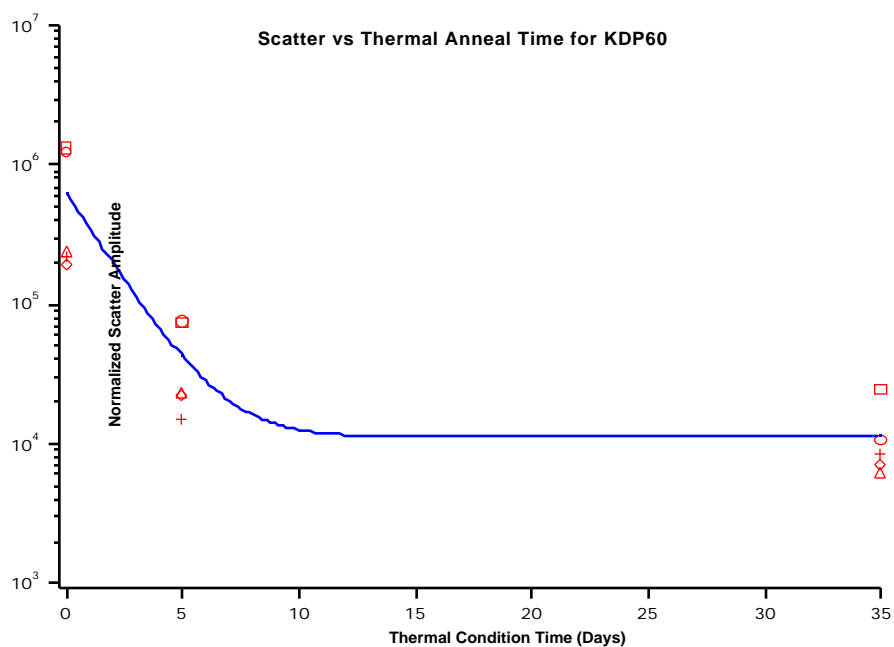


Figure 9. Normalized amplitude of linear features vs. thermal condition time in days at 160°C.

4. Conclusions

Optical scatter has not proven to be a useful technique for detecting where damage will initiate in KDP and DKDP. Numerous tests have shown that defects which cause point scattering are not typically the source from which damage initiates. The scatter diagnostic has proven to be very sensitive in detecting bulk damage in optics. Studies of our diagnostic have shown that we typically see all defects which cause point scattering irregardless of the direction viewed or from which direction the scatter beam is introduced into the sample. In looking at 90° side scattered light the true shape of a scatterer will not be evident using the diagnostic. Although these point scatterers do not typically initiate damage they sometimes do and, as a result, every effort should be made to get rid of them in the crystals by using constant filtration during the growth process.

While constant filtration does substantially reduce the number of point scatterers, recent tests have shown that it has done little to improve damage thresholds. Conventional wisdom is that constant filtration has been the reason for improved damage thresholds in conventionally grown crystals, however recent results have made us question whether constant filtration was indeed the reason for the improvement.

We believe linear scatter is due to index variations in the crystal parallel to the growth as a result of a change in lattice parameters. The location of linear features and damage sites seemed to coincide some of the time but a strong direct link could not be established. The amplitude of the linear scatter had no correlation with the damage.

Thermal conditioning of KDP was found to be a useful technique for increasing the damage threshold in regions which had high strain. Highly strained regions of crystals showed a dramatic decrease in the strain when thermally conditioned but regions with low strain showed little change. Linear scattering features of high amplitude showed a dramatic decrease in amplitude when thermally conditioned but low amplitude line features showed no change and we saw little effect of thermal conditioning on point scatterers.

5. Acknowledgments

We would like to thank Ron Vallene and Jose Vargas for optical finishing of the many samples that were used for this work. This work was performed under the auspices of the US department of Energy by Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

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